

# Developing probability distributions for transfer efficiencies for dermal exposure

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Many dermal exposure models use stochastic techniques to sample parameter distributions derived from experimental data to more accurately represent variability and uncertainty. Transfer efficiencies represent the fraction of a surface contaminant transferred from the surface to the skin during a contact event. Although an important parameter for assessing dermal exposure, examination of the literature confirms that no single study is large enough to provide a basis for a transfer efficiency distribution for use in stochastic dermal exposure models. It is therefore necessary to combine data sets from multiple studies to achieve the largest data set possible for distribution analysis. A literature review was conducted to identify publications reporting transfer efficiencies. Data sets were compared using the Kruskal–Wallis test to determine whether they arise from the same distribution. Combined data were evaluated for several theoretical distributions using the Kolmogorov–Smirnov and  $\chi^2$ -goodness-of-fit tests. Our literature review identified 35 studies comprising 25 different sampling methods, 25 chemicals, and 10 surface types. Distributions were developed for three different chemicals (chlorpyrifos, pyrethrin I, and piperonyl butoxide) on three different surface types (carpet, vinyl, and foil). Only the lognormal distribution was consistently accepted for each chemical and surface combination. Fitted distributions were significantly different (Kruskal–Wallis test;  $P < 0.001$ ) across chemicals and surface types. In future studies, increased effort should be placed on developing large studies, which more accurately represent transfer to human skin from surfaces, and on developing a normative transfer efficiency measure so that data from different methodologies can be compared.

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## Introduction

Human exposure analysis has traditionally been concerned with inhalation and dietary exposure routes. For certain populations, like young children, dermal exposure can be the most significant route of exposure to certain contaminants (Zartarian and Leckie, 1998). Relatively recently have dermal exposure analysis studies been published in the experimental literature (Fenske, 2000). The relevant dermal measurements that are published tend to be limited in scope and inconsistent from study to study. Not only is dermal exposure a difficult route to directly measure, it is also difficult to model. As concern for children's exposure in the residential environment grows, scientists and engineers continue to develop dermal exposure models that require experimentally derived parameters. Any resulting estimates from these models are limited by the quality of available input data sets.

Exposure model inputs must represent variability in time, space, and that between individuals in a population, and the uncertainty associated with both measurements from imperfect instruments and mathematical representations of complex physical, chemical, and biological processes (Frey and Cullen, 1995). Thus, the task of selecting single point estimates to represent these inputs obviously requires judgment. Modern dermal exposure models use stochastic techniques to sample from experimentally derived parameter distributions (e.g., environmental concentrations, transfer efficiencies, surface area, and so on) to more accurately represent the variability in the environment and the uncertainty of the measurements (Zartarian et al., 2000; Canales and Leckie, 2006; Zartarian et al., 2006).

Transfer efficiency, which represents the fraction of surface contaminant transferred to (or from) skin during a contact event (Cohen Hubal et al., 2000; Zartarian et al., 2000), may be one of the more important parameters when modeling dermal exposure (Xue et al., 2006), yet it is one of the most difficult to adequately (or accurately) measure. Part of the difficulty is that transfer efficiency may be a function of many parameters (e.g., physical nature of both surfaces; number, duration and pressure of contact; concentration on surface; time since the application of the chemical; temperature and humidity). In addition, there is inconsistency in experimental

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designs (e.g., methodologies, collection media, chemicals and surfaces) when measuring transfer efficiencies. For example, within the experimental literature there are several different studies that report transfer efficiencies; however, many of the variables, which may be important, such as temperature and humidity, are rarely controlled or reported. Therefore, it is nearly impossible to determine which variables have the greatest effect on transfer efficiencies. Nevertheless, distributions of this parameter are necessary for implementation of current stochastic dermal exposure models. In the experimental literature, there are many studies that report transfer efficiencies; however, many of the variables, such as temperature and humidity, are rarely controlled or reported. In lieu of more robust data from adequately designed experiments, transfer efficiency distributions are needed for dermal exposure modeling, and the current data that are available must be optimized.

The main objective of this study is to develop transfer efficiency distributions for use in current dermal exposure models. These models require chemical-specific distributions for transfer from smooth and textured surfaces (Zartarian et al., 2000; Canales and Leckie, 2006). This study will add to other standard exposure factor distributions (e.g., soil-to-skin adherence, hand-to-mouth frequency) in the current literature that were developed for use in stochastic exposure models (Finley et al., 1994; Burmaster and Crouch, 1997; Thompson, 1999; Xue et al., 2007).

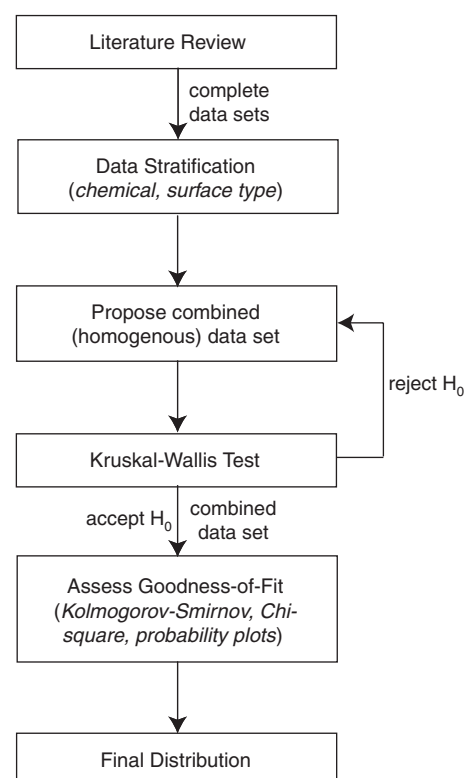
According to the United States Environmental Protection Agency (2000), distribution development should be conducted with data sets with more than 30 data points to reduce bias. If no single study in the reviewed literature is large enough ( $n > 30$ ) to provide the basis for parameter distribution evaluation, it will be necessary to combine experimental data from multiple studies to develop distributions for transfer efficiency. To use experimental data to develop input distributions for modeling, the data need to be critically evaluated for internal consistency and experimental uncertainty. In previous reviews of exposure factors, data sets were combined with respect to experimental design (Finley et al., 1994). The inconsistent experimental methods used to collect transfer efficiencies and lack of theoretical understanding of the important parameters driving transfer efficiency make combination of experimental data from multiple studies in a physically meaningful way (i.e., with respect to experimental design), very difficult. To achieve optimized data sets from the current experimental literature for determining the probability distributions, it may be necessary to combine data sets.

To fulfil the main objective of the current study, transfer efficiency distribution development, it was necessary to determine whether data in the current experimental literature could be optimized for distribution development by combining data from multiple studies using statistical tests. The goodness-of-fit (GOF) of the combined data to theoretical

distributions was evaluated to identify the best distributions for use in current models (Zartarian et al., 2000; Canales and Leckie, 2006). The experimental design of the combined data sets were explored retrospectively to determine whether there were any consistent trends in inclusion/exclusion of experimental methods, to provide insights and guidance for future experimental transfer efficiency studies.

## Materials and methods

As depicted in Figure 1, a literature review was first conducted to identify publications that report transfer efficiencies. Our literature review identified 35 studies directly reporting transfer efficiencies or adequate information to compute transfer efficiencies (Supplementary Table S-1). These studies included 25 different sampling methods, 25 chemicals, and 10 surface types. The majority of the studies report only mean values and were not included in our analysis because the mean alone does not provide information regarding the underlying distribution and constitutes an assumption of normality. We were unsuccessful in obtaining complete data sets from most of the authors, and the analysis is based upon those publications presenting primary data. Only 13 studies reported full experimental data sets. Four of the studies provided very little data on four different chemicals, and would not be able to be combined with other



**Figure 1.** Flow chart of transfer efficiency distribution development.

data sets. Therefore only nine studies were used to fit transfer efficiency distributions for three different chemicals (all pesticides) on four different surfaces with eight different methods, resulting in 78 distinct data sets. A brief description of each data set is provided in Table 1 (see the original references for complete study design). Note that not all studies report transfer efficiencies directly, but rather the initial mass on a surface and the mass transferred from the surface. For such studies, data on the mass of pesticide removed was divided by the amount applied to calculate transfer efficiencies.

Most of the experiments measured pesticide transfer efficiency from either carpet or vinyl. These two surfaces are assumed to represent textured and smooth surfaces, respectively, in current exposure models (Zartarian et al., 2000; Hore et al., 2006). Additional data were found measuring transfer efficiency from aluminum foil and turf. Although foil is not necessarily representative of the home environment, it may provide an estimate of maximum transfer efficiency, especially from non-porous residential surfaces such as glass or unpainted metal. It should be noted that most of the transfer efficiencies were measured relatively soon (within 24 h) after the pesticide application. Thus, reported values may be conservative estimates of transfer efficiency in the typical residential environment.

Several dislodgeable sampling devices have been developed as skin proxies to approximate transfer of a chemical from a contaminated surface to the skin, in the hopes of reducing chemical testing on humans. These methods include the Southwest Research Institute polyurethane foam (PUF) roller (Hsu et al., 1990), the California cloth roller (Ross et al., 1991), and the Dow drag sled (Vaccaro and Cranston, 1990). Studies have also been conducted with hand presses followed by subsequent hand wipes. Additionally, since children often have saliva moistened hands, saliva and other surrogate solutions (water, artificial saliva, and dioctyl sulfosuccinate) have been used to moisten the hands before measuring transfer efficiency from hand presses with surfaces.

All statistical tests were conducted with S-PLUS 6.0 (Insightful Corp., 2001). Data sets were compared using a non-parametric analysis of variance method (Figure 1), and Kruskal–Wallis test, to determine whether different combinations of data sets arise from the same distribution (Rice, 1995). The Kruskal–Wallis test was used as it does not make an assumption of normality and since data are replaced by ranks, outliers have less of an influence on the test, making it better suited for small data sets. Data sets were stratified by chemical and surface type (Figure 1), since previous studies have demonstrated that transfer efficiencies are chemical dependent (Cohen Hubal et al., 2005) and current dermal exposure models require transfer efficiency distributions for different surface types (Zartarian et al., 2000; Canales and Leckie, 2006; Hore et al., 2006). Given that all of the sampling methods are attempting to replicate chemical

transfer to human skin, the Kruskal–Wallis test was used to determine whether data sets from different sampling methods but for the same chemical and surface could be combined. Initially all of the data sets for a specific chemical and surface type were evaluated by Kruskal–Wallis test. Data sets were eliminated one by one, taking care to maximize the total number of data points until the Kruskal–Wallis *P*-value was greater than 0.05.

While empirical distribution functions (EDFs) are sometimes used in exposure modeling, we chose to develop theoretical distributions (e.g., normal, uniform, lognormal) for the combined transfer efficiency data sets. If a theoretical distribution can be found that fits the observed data reasonably well, it is generally preferable to using an EDF for the following reasons: (1) an EDF may have certain irregularities (particularly if the data set is small), which the theoretical distribution will “smooth out”; (2) it is not possible to generate values outside the range of observed values for the EDF; (3) a theoretical distribution is a more compact way of representing a set of data values making them easier to transport from model to model; and (4) it is easier to change a theoretical distribution for conducting a sensitivity analysis or adding new observations (Law and Kelton, 2000).

While selection of appropriate theoretical distribution models to test for GOF should start with a consideration of the underlying phenomena that generated the data (Frey and Cullen, 1995), it is not clear which processes influence transfer efficiencies. Therefore, several distributions were evaluated. Statistical GOF tests do not enable one to prove that an assumed distribution is correct. They only allow one to evaluate evidence that the model may be inadequate (Frey and Cullen, 1995).

Combined data were evaluated using both the Kolmogorov–Smirnov and  $\chi^2$ -GOF tests for normal, lognormal, exponential, gamma, Weibull, beta, and uniform distribution models (Figure 1). Individual distribution parameters were calculated according to Law and Kelton (2000). A description of the different parametric distributions and the equations used to calculate the parameters are presented in Supplementary Table S-2. The Kolmogorov–Smirnov GOF test compares a set of non-continuous data points with a given theoretical cumulative distribution function. The  $\chi^2$ -GOF test involves comparing the histogram of the data set and with a histogram derived from a given theoretical distribution. Two GOF tests were used because they both have limitations. Perhaps the largest liability of the Kolmogorov–Smirnov GOF test is that the parameters describing the theoretical distribution cannot be computed from the testing data set (Cullen and Frey, 1999). For large data sets, a method of cross-validation could be used, but for our purposes that is not possible because our data sets are too small. Therefore a less robust test, the  $\chi^2$ -GOF test, was also used for verification because the degrees of freedom can be

**Table 1.** Pesticide residue transfer efficiency data sets and experimental design<sup>a</sup>.

Study	Data set	Range	Name of study/summary	Reference
1	Nylon carpet; chlorpyrifos (0.5%) in aqueous spray			Camann et al. (1996)
	11	0.021–0.121	Cloth roller ( <i>n</i> = 6)	
	12	0.007–0.024	Drag sled ( <i>n</i> = 6)	
	13	0.005–0.018	PUF roller ( <i>n</i> = 6)	
	Level-loop polypropylene carpet; chlorpyrifos (0.5%)			
	14	0.016–0.041	Cloth roller ( <i>n</i> = 6)	
	15	0.010–0.031	Drag sled ( <i>n</i> = 6)	
	16	0.009–0.029	PUF roller ( <i>n</i> = 6)	
	Nylon carpet; chlorpyrifos (0.5%) in aqueous spray			
	17	0.002–0.007	Dry contact medium, drag sled ( <i>n</i> = 4)	
	18	0.002–0.003	Dry contact medium: PUF roller ( <i>n</i> = 4)	
	19	0.002–0.018	Moistened contact medium, drag sled ( <i>n</i> = 5)	
	110	0.003–0.033	Moistened contact medium, PUF roller ( <i>n</i> = 5)	
	Hand presses after two compounds had been applied to foil			
	111	0.77–0.96	Chlorpyrifos ( <i>n</i> = 12)	
	112	0.71–0.94	Pyrethrin I ( <i>n</i> = 12)	
	Plush nylon carpet; mixed aqueous spray			
	113	0.0003–0.002	Chlorpyrifos (0.25%), drag sled ( <i>n</i> = 6)	
	114	0.0001–0.0008	Chlorpyrifos (0.25%), PUF roller ( <i>n</i> = 6)	
	115	0.00001–0.00003	Chlorpyrifos (0.25%), hand press ( <i>n</i> = 3)	
	116	0.0005–0.002	Piperonyl butoxide (0.25%), drag sled ( <i>n</i> = 6)	
	117	0.0002–0.0009	Piperonyl butoxide (0.25%), PUF roller ( <i>n</i> = 6)	
	118	0.00002–0.00005	Piperonyl butoxide (0.25%), hand press ( <i>n</i> = 4)	
	119	0.0006–0.005	Pyrethrin I (0.25%), drag sled ( <i>n</i> = 6)	
	120	0.0003–0.0008	Pyrethrin I (0.25%), PUF roller ( <i>n</i> = 6)	
	Plush nylon carpet; mixed aerosol			
	121	0.010–0.056	Piperonyl butoxide (1%), drag sled ( <i>n</i> = 6)	
	122	0.013–0.019	Piperonyl butoxide (1%), PUF roller ( <i>n</i> = 6)	
	123	0.001–0.009	Piperonyl butoxide (1%), hand press ( <i>n</i> = 4)	
	124	0.008–0.070	Pyrethrin I (0.2%), drag sled ( <i>n</i> = 6)	
	125	0.009–0.021	Pyrethrin I (0.2%), PUF roller ( <i>n</i> = 6)	
	Sheet vinyl; mixed formulation			
	126	0.103–0.601	Chlorpyrifos (0.25%) drag sled	
	127	0.030–0.242	Chlorpyrifos (0.25%), PUF roller ( <i>n</i> = 6)	
	128	0.005–0.091	Chlorpyrifos (0.25%), hand press ( <i>n</i> = 18)	
	129	0.087–0.571	Piperonyl butoxide (0.25%), drag sled ( <i>n</i> = 6)	
	130	0.024–0.278	Piperonyl butoxide (0.25%), PUF roller ( <i>n</i> = 6)	
	131	0.006–0.128	Piperonyl butoxide (0.25%), hand press ( <i>n</i> = 18)	
	132	0.096–0.449	Pyrethrin I (0.25%), drag sled ( <i>n</i> = 6)	
	133	0.040–0.216	Pyrethrin I (0.25%), PUF roller ( <i>n</i> = 6)	
	134	0.002–0.130	Pyrethrin I (0.25%), hand press ( <i>n</i> = 18)	
2	Moistened hand presses; nylon carpet			Camann et al. (1995)
	21	0.004–0.009	Chlorpyrifos (0.25%), artificial saliva ( <i>n</i> = 6)	
	22	0.007–0.023	Chlorpyrifos (0.25%), DSS ( <i>n</i> = 6)	
	23	0.006–0.021	Chlorpyrifos (0.25%), human saliva ( <i>n</i> = 6)	
	24	0.017–0.054	Pyrethrin (0.025%), artificial saliva ( <i>n</i> = 6)	
	25	0.023–0.079	Pyrethrin (0.025%), DSS ( <i>n</i> = 6)	
	26	0.015–0.078	Pyrethrin (0.025%), human saliva ( <i>n</i> = 6)	
	27	0.009–0.027	Piperonyl butoxide (0.25%), artificial saliva ( <i>n</i> = 6)	
	28	0.016–0.054	Piperonyl butoxide (0.25%), DSS ( <i>n</i> = 6)	
	29	0.004–0.009	Piperonyl butoxide (0.25%), human saliva ( <i>n</i> = 6)	
3	Sheet vinyl; broadcast spray; dried for 4 h			Clothier (2000)
	31	0.007–0.026	Chlorpyrifos(0.25%), dry hand press ( <i>n</i> = 6)	
	32	0.015–0.082	Chlorpyrifos(0.25%), water-wetted hand press ( <i>n</i> = 6)	
	33	0.018–0.097	Chlorpyrifos(0.25%), saliva-wetted hand press ( <i>n</i> = 6)	
	34	0.012–0.076	Chlorpyrifos(0.25%), PUF roller ( <i>n</i> = 6)	
	35	0.010–0.060	Pyrethrin I (0.025%), dry hand press ( <i>n</i> = 6)	
	36	0.031–0.192	Pyrethrin I (0.025%), water-wetted hand press ( <i>n</i> = 6)	

Table 1. Continued

Study	Data set	Range	Name of study/summary	Reference
3. Continued	37	0.055–0.177	Pyrethrin I (0.025%), saliva-wetted hand press ( <i>n</i> = 6)	Clotheir (2000)
	38	0.019–0.106	Pyrethrin I (0.025%), PUF roller ( <i>n</i> = 6)	
	39	0.006–0.025	Piperonyl butoxide (0.25%) Dry hand press ( <i>n</i> = 6)	
	310	0.012–0.085	Piperonyl butoxide (0.25%), water-wetted hand press ( <i>n</i> = 6)	
	311	0.018–0.091	Piperonyl butoxide (0.25%), saliva-wetted hand press ( <i>n</i> = 6)	
	312	0.009–0.085	Piperonyl butoxide (0.25%), PUF roller ( <i>n</i> = 6)	
4	Carpet; mixed formulation			Fortune (1997a)
	41	0.005–0.029	Chlorpyrifos (0.05%), PUF roller ( <i>n</i> = 21)	
	42	0.013–0.098	Chlorpyrifos (0.05%), cloth roller ( <i>n</i> = 21)	
	43	0.005–0.042	Chlorpyrifos (0.05%), drag sled ( <i>n</i> = 21)	
	44	0.006–0.086	Pyrethrin I (0.5%), PUF roller ( <i>n</i> = 21)	
	45	0.013–0.073	Pyrethrin I (0.5%), cloth roller ( <i>n</i> = 21)	
	46	0.008–0.083	Pyrethrin I (0.5%), drag sled ( <i>n</i> = 21)	
	47	0.007–0.039	Piperonyl butoxide (0.05%), PUF roller ( <i>n</i> = 21)	
	48	0.020–0.195	Piperonyl butoxide (0.05%), cloth roller ( <i>n</i> = 21)	
	49	0.008–0.065	Piperonyl butoxide (0.05%), drag sled ( <i>n</i> = 21)	
5	Turf; commercial aqueous mixture chlorpyrifos (0.17%)			Fortune (1997b)
	51	0.0004–0.001	PUF roller ( <i>n</i> = 7)	
	52	0.0002–0.001	Drag sled ( <i>n</i> = 7)	
6	Foil; 0.10 µg/µl of chlorpyrifos and 1.4 µg/µl of pyrethrin I			Geno et al. (1996)
	61	0.80–0.96	Chlorpyrifos, subject A ( <i>n</i> = 6)	
	62	0.77–0.94	Chlorpyrifos, subject B ( <i>n</i> = 6)	
	63	0.74–0.94	Pyrethrin I, subject A ( <i>n</i> = 6)	
	64	0.71–0.93	Pyrethrin I, subject B ( <i>n</i> = 6)	
7	Foil; dried 90 s			Hsu et al. (1990)
	71	0.055–0.088	PUF Roller, chlorpyrifos ( <i>n</i> = 3)	
	72	0.061–0.11	Human hand heel, chlorpyrifos ( <i>n</i> = 6)	
8	Nylon carpet; foggers containing chlorpyrifos (1.000%)			Krieger et al. (2000)
	81	0.006–0.201	Cloth roller ( <i>n</i> = 12)	
9	Foggers containing chlorpyrifos (0.5%); cloth roller			Ross et al. (1991)
	91	0.029–0.258	Stain-resistant carpet ( <i>n</i> = 6)	
	92	0.025–0.055	Nylon carpet ( <i>n</i> = 6)	
	Facility carpet; foggers containing chlorpyrifos (0.5%); cloth roller			
	93	0.011–0.045	0 h Post application ( <i>n</i> = 4)	
	94	0.006–0.025	6 h Post application ( <i>n</i> = 4)	
	95	0.006–0.024	12 h Post application ( <i>n</i> = 4)	

<sup>a</sup>Underlining indicates data set included (Kruskal–Wallis  $P$ -value  $> 0.05$ ).

adjusted for the number of distribution parameters calculated from the testing data set. As depicted in Figure 1, probability plots were used to confirm the results (Rice, 1995).

## Results

Examination of the literature on transfer efficiency confirms that no single study is large enough ( $n > 30$ ) to provide the basis for parameter distribution evaluation (US EPA, 2000), and it is necessary to pool data from multiple studies using

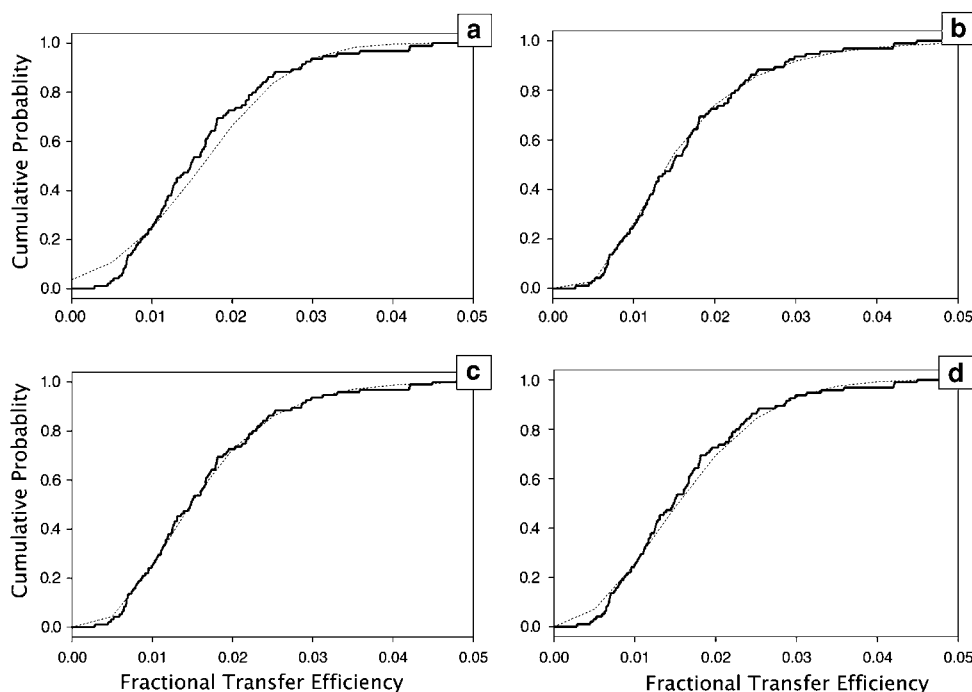
Kruskal–Wallis test. A summary of the data sets that were combined, the number of data points and the Kruskal–Wallis  $P$ -value are given in Table 2. Two data sets were found that reported transfer efficiency from turf, but the data sets could not be joined and they were not individually large enough to fit a distribution.

The experimental methodologies of the combined data sets were retrospectively reassessed to explore whether there were any consistent trends in inclusion/exclusion of experimental methods. The individual data sets that were included in the aggregate data sets (i.e., excluded) because they passed the

**Table 2.** Summary of Kolmogorov–Smirnov and  $\chi^2$ -GOF tests<sup>a</sup>.

Chemical	Surface	Data sets used	Kruskal–Wallis <i>P</i> -value	<i>n</i>	Distribution	Parameters	Kolmogorov–Smirnov <i>P</i> -value	$\chi^2$ -test <i>P</i> -value
Chlorpyrifos	Carpet	12, 13, 15, 16, 110, 22, 23, 41, 43, 93, 94, 95	0.089	95	Normal	$\hat{\mu} = 0.0162, \hat{\sigma} = 0.009$	<b>0.2817</b>	<b>0.1469</b>
					Lognormal	$\hat{\mu} = -4.26, \hat{\sigma} = 0.54$	<b>0.9282</b>	<b>0.4936</b>
					Exponential	$\hat{\beta} = 0.0162$	0	0
					Gamma	$\hat{\alpha} = 3.759, \hat{\beta} = 0.004$	<b>0.9979</b>	<b>0.6247</b>
					Beta	$\hat{\alpha}_1 = 0.85, \hat{\alpha}_2 = 42.126$	0	0
					Weibull	$\hat{\alpha} = 2.008, \hat{\beta} = 0.018$	<b>0.6784</b>	<b>0.5715</b>
	Vinyl	127, 128, 32, 33, 34	0.0879	42	Uniform	$\hat{a} = 0.003, \hat{b} = 0.045$	0	0
					Normal	$\hat{\mu} = 0.052, \hat{\sigma} = 0.050$	<b>0.1372</b>	0.0002
					Lognormal	$\hat{\mu} = -3.301, \hat{\sigma} = 0.845$	<b>0.97</b>	<b>0.5809</b>
					Exponential	$\hat{\beta} = 0.052$	<b>0.2559</b>	<b>0.4439</b>
					Gamma	$\hat{\alpha} = 1.647, \hat{\beta} = 0.031$	<b>0.8751</b>	<b>0.5256</b>
					Beta	$\hat{\alpha}_1 = 2.898, \hat{\alpha}_2 = 45.901$	0.0069	0.0044
	Foil	111, 61, 62	0.1975	24	Weibull	$\hat{\alpha} = 1.25, \hat{\beta} = 0.06$	<b>0.5567</b>	0.1509
					Uniform	$\hat{a} = 0.005, \hat{b} = 0.242$	0	0
					Normal	$\hat{\mu} = 0.866, \hat{\sigma} = 0.066$	<b>0.2478</b>	<b>0.1562</b>
					Lognormal	$\hat{\mu} = -0.147, \hat{\sigma} = 0.076$	<b>0.2488</b>	<b>0.3766</b>
					Exponential	$\hat{\beta} = 0.886$	0	0
					Gamma	$\hat{\alpha} = 25.166, \hat{\beta} = 0.034$	0.0099	0.0022
	Turf	51, 52	0.004	24	Beta	$\hat{\alpha}_1 = 8.277, \hat{\alpha}_2 = 1.374$	<b>0.1778</b>	0.0022
					Weibull	$\hat{\alpha} = 14, \hat{\beta} = 0.89$	<b>0.1832</b>	0.0068
					Uniform	$\hat{a} = 0.769, \hat{b} = 0.965$	<b>0.2323</b>	<b>0.1562</b>
					None of the data sets were large enough to fit a distribution			
Pyrethrins I	Carpet	124, 125, 24, 26, 44, 46	0.0547	66	Normal	$\hat{\mu} = 0.027, \hat{\sigma} = 0.020$	0.0035	0
					Lognormal	$\hat{\mu} = -3.864, \hat{\sigma} = 0.675$	<b>0.3931</b>	<b>0.265</b>
					Exponential	$\hat{\beta} = 0.027$	0.0019	0.0012
					Gamma	$\hat{\alpha} = 2.253, \hat{\beta} = 0.012$	<b>0.0976</b>	0.0038
					Beta	$\hat{\alpha}_1 = 0.85, \hat{\alpha}_2 = 42.126$	0	0
					Weibull	$\hat{\alpha} = 1.47, \hat{\beta} = 0.03$	<b>0.1162</b>	0.0011
	Vinyl	134, 35, 38	0.147	30	Uniform	$\hat{a} = 0.006, \hat{b} = 0.086$	0	0
					Normal	$\hat{\mu} = 0.037, \hat{\sigma} = 0.030$	<b>0.437</b>	<b>0.197</b>
					Lognormal	$\hat{\mu} = -3.66, \hat{\sigma} = 0.964$	<b>0.9554</b>	<b>0.5304</b>
					Exponential	$\hat{\beta} = 0.037$	<b>0.4695</b>	<b>0.3397</b>
					Gamma	$\hat{\alpha} = 1.546, \hat{\beta} = 0.024$	<b>0.975</b>	<b>0.6083</b>
					Beta	$\hat{\alpha}_1 = 0.407, \hat{\alpha}_2 = 8.232$	0.0044	0.0001
	Foil	112, 63, 64	0.4241	24	Weibull	$\hat{\alpha} = 1.28, \hat{\beta} = 0.04$	<b>0.9629</b>	<b>0.6083</b>
					Uniform	$\hat{a} = 0.002, \hat{b} = 0.130$	0.0001	0.0004
					Normal	$\hat{\mu} = 0.831, \hat{\sigma} = 0.079$	<b>0.7014</b>	<b>0.0584</b>
					Lognormal	$\hat{\mu} = -0.188, \hat{\sigma} = 0.096$	<b>0.7271</b>	<b>0.0584</b>
					Exponential	$\hat{\beta} = 0.831$	0	0
					Gamma	$\hat{\alpha} = 25.166, \hat{\beta} = 0.033$	<b>0.0984</b>	0.0204
Piperonyl butoxide	Carpet	122, 27, 28, 47, 49	0.0539	60	Beta	$\hat{\alpha}_1 = 42.104, \hat{\alpha}_2 = 7.908$	<b>0.0827</b>	0.0022
					Weibull	$\hat{\alpha} = 12, \hat{\beta} = 0.87$	<b>0.561</b>	<b>0.0584</b>
					Uniform	$\hat{a} = 0.710, \hat{b} = 0.942$	<b>0.3686</b>	<b>0.0584</b>
					Normal	$\hat{\mu} = 0.021, \hat{\sigma} = 0.012$	<b>0.0961</b>	0.0081
					Lognormal	$\hat{\mu} = -4.001, \hat{\sigma} = 0.513$	<b>0.9528</b>	<b>0.6472</b>
					Exponential	$\hat{\beta} = 0.021$	0.0001	0
	Vinyl	131, 39, 310, 311, 312	0.1437	42	Gamma	$\hat{\alpha} = 3.809, \hat{\beta} = 0.006$	<b>0.1239</b>	0.0233
					Beta	$\hat{\alpha}_1 = 0.85, \hat{\alpha}_2 = 42.126$	0	0
					Weibull	$\hat{\alpha} = 1.85, \hat{\beta} = 0.02$	<b>0.0812</b>	<b>0.0899</b>
					Uniform	$\hat{a} = 0.007, \hat{b} = 0.065$	0	0
					Normal	$\hat{\mu} = 0.036, \hat{\sigma} = 0.026$	<b>0.0994</b>	0.0001
					Lognormal	$\hat{\mu} = -3.625, \hat{\sigma} = 0.808$	<b>0.944</b>	<b>0.8604</b>
					Exponential	$\hat{\beta} = 0.036$	<b>0.3156</b>	<b>0.4439</b>
					Gamma	$\hat{\alpha} = 1.849, \hat{\beta} = 0.019$	<b>0.8262</b>	<b>0.2952</b>
					Beta	$\hat{\alpha}_1 = 0.407, \hat{\alpha}_2 = 8.232$	0.0009	0
					Weibull	$\hat{\alpha} = 1.41, \hat{\beta} = 0.04$	<b>0.5669</b>	<b>0.4729</b>
					Uniform	$\hat{a} = 0.005, \hat{b} = 0.091$	0.001	0.0015

<sup>a</sup>Parameter calculations and notations for each distribution are summarized in Table S-1 (Supplementary Material).

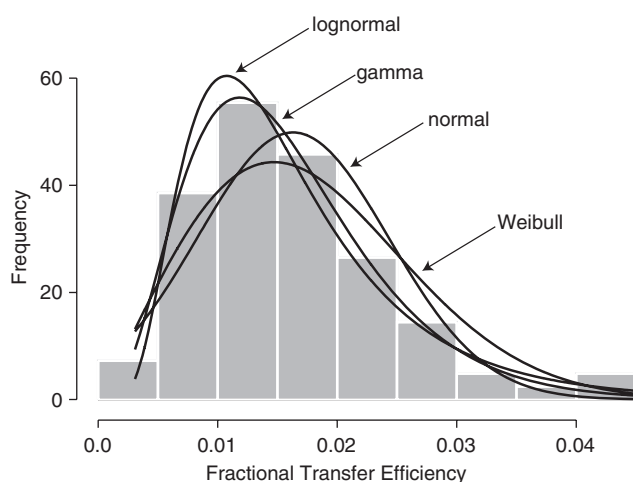


**Figure 2.** Comparison of transfer efficiency EDF for chlorpyrifos from carpet ( $n = 83$ ) with theoretical CDFs, normal (a), lognormal (b), gamma (c), and Weibull (d).

Kruskal–Wallis test are underlined in Table 1. Although data sets were combined across different experimental designs, no consistent trends were observed with respect to exclusion of PUF roller, Dow drag sled, California cloth roller, and different types of hand presses. Data sets collected from dry and a variety of moistened hand presses were combined, perhaps indicating that skin moisture may not affect transfer efficiency. Data sets were combined with different application concentrations and formulations, which may indicate that these variables may not affect transfer efficiency significantly. Data sets sampled at different times after application of the pesticide were also combined, demonstrating that transfer efficiency may not decrease with time. Five experiments (data sets 113 to 120 from Hsu et al., 1990; Camann et al., 1996; Fortune, 1997b; Krieger et al., 2000; and data sets 91–92 from Ross et al., 1991) were completely excluded, indicating that data from these experiments were statistically different from those from other experiments. Since sampling methods for these experiments are similar to others that were combined, it is not clear what variation in experimental design or other variables can account for these differences. This analysis demonstrates the need for further consistency when collecting transfer efficiency data, where variables such as sampling methods, application formulation and concentration, time following application, and environmental conditions are explored in a systematic manner, perhaps using a chamber where all of the variables can be controlled (Johnson et al., 2006).

Figures 2 and 3 provide a visual example of the parametric fits for both the CDFs and the PDFs for transfer efficiency of chlorpyrifos from carpet. The results of both the Kolmogorov–Smirnov and  $\chi^2$ -GOF tests are summarized in Table 2. While several distributions were assessed, only the null hypotheses for the lognormal distribution were consistently accepted for each chemical and surface combination for both GOF tests. Even though two combinations of data did have a larger  $P$ -value with gamma or beta distribution, they too can be fit by a lognormal distribution. This may suggest that the same underlying physical processes that govern the other transfer efficiency data sets resulting in lognormal distribution, also govern these data sets. Probability plots were constructed to confirm the results of each GOF test. The combined data sets did plot linearly for each of the distributions not rejected, indicating GOF between the combined data sets and theoretical distributions. No changes in slope or curvature were observed as a result of combining multiple data sets, demonstrating that the data points did belong to the same underlying distribution.

The lognormal distributions for the three chemicals for each surface type (Table 3; Figure 4) were evaluated by Kruskal–Wallis test. The Kruskal–Wallis  $P$ -value for all surface and chemical combinations was less than 0.0001, indicating that distributions are statistically different. Within each chemical, a trend is apparent for each of the surface types, with the most pesticide transferring from contacts with foil (if available), followed by vinyl and carpet (Figure 4).



**Figure 3.** Histogram of transfer efficiency of chlorpyrifos from carpet and parametric fits.

**Table 3.** Lognormal distributions for modeling transfer efficiencies (fraction)<sup>a</sup>.

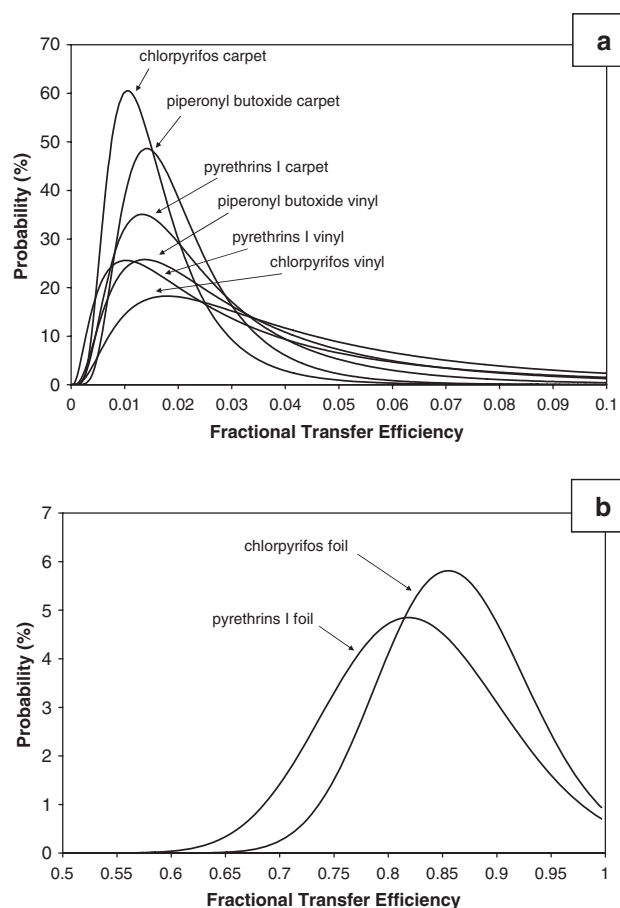
Chemical	Surface	$\hat{\mu}$	$\hat{\sigma}$	GM	GSD
Chlorpyrifos	Carpet	-4.26	0.54	0.01	1.70
	Vinyl	-3.30	0.85	0.04	2.34
	Foil	-0.15	0.08	0.86	1.08
Pyrethrins I	Carpet	-3.86	0.68	0.02	1.97
	Vinyl	-3.66	0.96	0.03	2.61
	Foil	-0.19	0.10	0.83	1.11
Piperonyl Butoxide	Carpet	-4.00	0.51	0.02	1.67
	Vinyl	-3.63	0.81	0.03	2.25

<sup>a</sup>Distributions should be truncated at 1.0.

For instance, for chlorpyrifos the geometric means (and geometric standard deviations) for transfer efficiency were calculated as 0.86 (1.08), 0.04 (2.34), and 0.01 (1.70) for foil, vinyl, and carpet, respectively. Until better data are available for reassessing transfer efficiency probability distributions, the lognormal distributions presented in Table 3 should be used for modeling purposes. Since transfer efficiency cannot exceed 1.0, the recommended distributions should be truncated when used in models. Caution should be used when extending these distributions to other chemicals and surface types.

## Discussion

Whereas transfer efficiency data were available for 25 different chemicals, multiple data sets were only available for three chemicals. These three chemicals are pesticides often used to treat indoor infestations of fleas. The 1996 Food Quality Protection Act and concern for children's residential pesticide exposure have driven most of the work attempting



**Figure 4.** Recommended probability distribution functions of transfer efficiencies from (a) carpet, vinyl, and (b) foil.

to estimate the transfer efficiency of these pesticides. Additional research needs to be conducted to determine transfer efficiencies for other chemicals (e.g., polybrominated diphenyl ethers, phthalates, perfluorooctanesulfonate) and the complete data sets should be made available for distribution development.

Probability distributions for transfer efficiency of chlorpyrifos from carpet and vinyl have been published as part of EPA's Residential Stochastic Human Exposure and Dose Simulation Model for Pesticides (Residential-SHEDS) (Zartarian et al., 2000). The probability distributions for SHEDS were developed from some of the data sets used in the current study and additional data sets that were rejected from this study because only summary statistics were available. Zartarian et al. (2000) also reported that the transfer efficiency of chlorpyrifos from carpet was best represented by a lognormal distribution ( $n=24$ , geometric mean = 0.32%, geometric standard deviation = 4.12%). The researchers did not have enough data points ( $n=4$ ) to fit a distribution for the transfer efficiency from vinyl, and hence they used a uniform distribution (minimum = 0.7%,



maximum = 10%). Although on the same order of magnitude as the distributions fit in the current study, the distribution parameters developed by Zartarian et al. (2000) are a bit lower (geometric mean of 1% in this study compared with 0.32%; Zartarian et al. (2000)) and the resulting distribution is statistically different (Kruskal–Wallis  $P$ -value  $< 0.001$ ). Since different data sets were used, this indicates that it is important to be careful while selecting data sets to combine, as it will affect the fit distribution.

Clearly, additional data are needed to verify the accuracy of model input parameter distributions. Future studies need to be designed to provide large data sets that are systematically collected, thereby reducing uncertainty and providing a more accurate representation of the variability of transfer efficiencies. In forthcoming experiments, further analysis should be conducted to determine which variables (e.g., chemical properties, physical properties of the surface, time after application, pesticide concentration, temperature, humidity, and multiple contacts) are important in determining transfer efficiency. While transfer efficiency appears to vary with chemical compound (Cohen Hubal et al., 2005), additional work should be conducted to determine differences in transfer efficiency due to chemical characteristics and to identify non-toxic chemicals that could serve as surrogates for pesticides in transfer efficiency studies. Furthermore, increased effort should be placed on developing studies that more accurately represent transfer from surfaces to human skin, and on constructing a normative measure so that data from different methodologies can be compared or transformed to represent realistic surface-to-skin transfer. If sufficient experimental data are collected for a variety of chemicals and surfaces, an empirical model can be developed as a function of the variables that contribute most to dermal transfer efficiency, thus enabling estimates for a wider range of chemicals and exposure scenarios. It is important that the complete data sets from future transfer efficiency studies be made available for distributional development for stochastic dermal exposure modeling.

As shown under Results, it is not possible to combine data sets according to experimental design as it is not clear which variables are important. The current study does show how it is possible to combine data sets that have the same underlying distribution, as evident by lack of deviations on the probability plots, by using statistical tests. The combined data sets provide a much larger number of data points that can improve the fit of theoretical distributions and reduce bias by increasing the degrees of freedom. If the complete data sets, instead of summary statistics, for the other transfer efficiency studies become available, the subsequent data points would allow a more rigorous test of the distributional fit. Results of this evaluation underscore the difficulty of fitting distributions for transfer efficiencies due to small sample size, differences in experimental methodologies, and inaccessibility of complete data sets. These statistical methods

could be used to develop distributions for other exposure factors.

Given the need for dermal exposure estimates as part of the risk assessment process, dermal exposure models have progressed more rapidly than the experimental literature that provides the input parameter values for these models. At this time, the available transfer efficiency data sets are relatively small and many of the current publications present only summary statistics, thus limiting distribution development for use in stochastic models. A data repository for transfer efficiency experimental data sets would be helpful in creating larger available data sets. As these larger data sets and superior methodologies and experimental designs become available, they can be used to develop new distributions for dermal exposure models. Increased quality in experimental data used to develop parameter distributions will decrease the uncertainty associated with these distributions, resulting in improved dermal exposure estimates. Improved estimates of dermal exposure may also improve non-dietary ingestion exposure estimates from hand-to-mouth contacts.

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